OFFICE OF NAVAL RESEARCH

FINAL REPORT

PUBLICATIONS/PATENTS/PRESENTATIONS/HONORS/STUDENTS REPORT

for

GRANT: N00014-00-1-0371

PR Number

Modification of Mossbauer Spectra by Laser Radiation

Principal Investigator: Olga Kocharovskaya

Texas A&M University

College Station, TX 77802

Co-Investigator: Zameer Hasan

Department of Physics, Temple University

Philadelphia, PA 19122

Co-Investigator: James Carroll

Department of Physics and Astronomy
Center for Photon-Induced Processes X-Ray Effects
Youngstown State University
One university Plaza
Youngstown, Ohio 44555-3616

OFFICE OF NAVAL RESEARCH
PUBLICATIONS/PATENTS/PRESENTATIONS/HONORS REPORT

PR Number:
Contract/Grant Number: N00014-00-1-0371
Contract/Grant Title: Modification of Mossbauer Spectra by Laser Radiation

Principal Investigator: Olga Kocharovskaya
Mailing Address: Department of Physics, Texas A&M University; College Station, TX 77843-4242

Phone number: (979)845-2012 Fax Number: (979)458-1235
Email address: kochar@atlantic.tamu.edu

Co-Investigator: Zameer Hasan
Mailing Address: Department of Physics, Temple University, Philadelphia, PA 19122

Phone number: (215) 204-7648 Fax Number: (215) 204-5652
Email address: zhasan@astro.temple.edu http address:

Co-Investigator: James Carroll
Mailing Address: Department of Physics and Astronomy, Center for Photon-Induced Processes X-Ray Effects, Youngstown State University, One university Plaza, Youngstown, Ohio 44555-3616

Phone number: (330) 742-3617 Fax Number: (330) 742-3121
Email address: jjcarroll@cc.ysu.edu http address:

A Number of Papers submitted to refereed journals, but not published: 10

[1] R.Kolesov and O. Kocharovskaya, Ultrashort pulses generation in solid media with a long lived spin coherence, Phys. Rev.A., submitted.

[2] R. Coussement, Y. Rostovtsev, J. Odeurs, P. Mandel, R. Shakhmuratov, and O. Kocharovskaya, EIT for gamma radiation via nuclear level-crossing, Phys. Rev. Lett., submitted.

[3] Y. Rostovtsev. O. Kocharovskaya, Laser-Mossbauer spectroscopy as a new tool for nuclear transitions. Hyperfine Interaction, submitted.

[4] O.Kocharovskaya, R.Kolesov and Yu.Rostovtsev, Mossbauer gamma-ray laser with coherent optical driving, Hyperfine interactions, submitted.

[5] O. Kocharovskaya, A. Belyanin, and Y. Rostovtsev, Atomic and Nuclear Interference Effects for Quantum Information Processing, Quantum Computers and Computing, accepted for publication (2002).

[6] Y.V. Radionychev, M.D. Tokman, A.G. Litvak, O. Kocharovskaya, Vibrationally induced transparency in optical dense resonance medium, Phys. Rev. Let., submitted.

[7] A.A. Belyanin, V.V. Kocharovsky, Vl.V. Kocharovsky, M.O. Scully, Transistor laser for wave mixing. Phys. Rev. A, submitted.

[8] A. Javan, O. Kocharovskaya, H. Lee, M. O. Scully, Narrowing of electromagnetically induced transparency resonance in a Doppler broadened medium, Phys. Rev. A, submitted.

[9] E. Kuznetsova, P. Hemmer, O. Kocharovskaya, M.O. Scully, "Effects of inhomogeneous line broadening on Electromagnetically Induced Transparency (EIT) and slow group velocity," Phys. Rev. A. submitted.

[10] Y. Rostovtsev. O. Kocharovskaya, G. Welch, M.O. Scully, Slow, ultra-slow and freezing light. Optics Photonics News, submitted.

B Number of Papers published in refereed journals (for each, provide a complete citation): 21

[1] R. Kolesov, Y. Rostovtsev, O. Kocharovskaya, Laser control of Mossbauer spectra as a way to gamma-ray lasing, Optics Communications, 179/1-6, 25 May (2000).

[2] A. B. Matsko, Y. V. Rostovtsev, H. Z. Cummins, M. O. Scully, Using slow light to enhance acousto-optical effects: Application to squeezed light, Phys. Rev. Lett. 84, 5752 (2000).

[3] H. Lee, Yu. Rostovtsev, M. Scully, Asymmetries between absorption and stimulated emission in driven three-level systems, Phys. Rev. A 62, 063804 (2000).

[4] A.I. Artemiev, M.V. Fedorov, Yuri V. Rostovtsev, Gershon Kurizki, and Marlan O. Scully, Free electron laser without inversion: gain optimization and implementation scheme, Phys. Rev. Lett. 85, 4510 (2000).

[5] O.Kocharovskaya, Y.Rostovtsev, M.O.Scully, Freezing light via atomic coherence, Phys. Rev. Lett., 86, 628 (2001).

[6] M.O. Scully, G.S. Agarwal, O. Kocharovskaya, V.V. Kozlov, A.B. Matsko,"Mixed electromagnetically and self-induced transparency", Opt. Express 8, 66 (2001).

[7] R. Kolesov, E. Kuznetsova, Possibility of polarization-selective optical pumping of nuclei in solids and its detection in the Mossbauer spectra, Phys. Rev. B63, 184107 (2001).

[8] Belyanin A.A., Capasso F., Kocharovsky V.V., Kocharovsky Vl.V., Scully M.O., Infrared generation in low-dimensional semiconductor heterostructures via quantum coherence, Phys. Rev. A, 2001, 63, 053803.

[9] Belyanin A.A., Bentley C., Capasso F., Kocharovsky V.V., Kocharovsky Vl.V., Scully M.O., Inversionless lasing with self-generated driving field, Phys. Rev. A, 2001, 64, 013814.

[10] Belyanin A.A., Capasso F., Kocharovsky V.V., Kocharovsky Vl.V., Pestov D.S., Scully M.O., Resonant parametric generation of infrared radiation on intersubband transitions in low-dimensional semiconductor heterostructures, Nanotechnology, 2001, 12, 1.

[11] R. Kolesov, Optical continua generation in a coherently prepared Raman medium, Phys. Rev. A 64, 063819 (2001)

[12] Y. V. Rostovtsev, G. Kurizki, and M. O. Scully, Broadband optical gain via interference in the free electron laser: principles and proposed realizations, Phys. Rev. E64, 026501 (2001).

[13] Y. V. Rostovtsev, A. B. Matsko, R. Shelby, M. O. Scully, Phase-matching condition between acoustic and optical waves in doped fibers, Optics and Spectroscopy 91, 519 (2001).

[14] A.B. Matsko, Yu. Rostovtsev, O. Kocharovskaya, A. Zibrov, M.O. Scully, Nonadiabatic Approach to Quantum Optical Information Storage, Phys. Rev. A 64, 043809 (2001).

[15] Y. Rostovtsev, and O. Kocharovskaya, Modification of Mossbauer spectra under the action of electromagnetic fields, Hyperfine Interaction 135, 233-255 (2001).

[16] A.B. Matsko, Yu. Rostovtsev, O. Kocharovskaya, A. Zibrov, M.O. Scully, Nonadiabatic Approach to Quantum Optical Information Storage, Phys. Rev. A 64, 043809 (2001).

[17] Belyanin A.A., Capasso F., Kocharovsky V.V., Kocharovsky Vl.V., Coherent radiation from neutral molecules moving above the grating, Phys. Rev. Lett. 88, 053602 (2002).

[18] A. B. Matsko, Y. V. Rostovtsev, M. Fleischhauer, and M. O. Scully, Anomalous stimulated Brillouin scattering via ultraslow light, Phys. Rev. Lett. **86**, 2006 (2001).

[19] O. Kocharovskaya, A.B. Matsko, Y. Rostovtsev, Lasing without inversion via decay induced coherence, Phys. Rev. A65, 013803 (2002).

[20] A. S. Zibrov, A. B. Matsko, O. Kocharovskaya, Y. V. Rostovtsev, G. R. Welch, and M. O. Scully, Transporting and Time Reversing Light via Atomic Coherence, Phys. Rev. Lett. 88, 103601 (2002).

[21] C. Y. Ye, A. S. Zibrov, Yu. V. Rostovtsev, and M. O. Scully, Unexpected Doppler-free Resonance in Generalized Double Dark States, Phys. Rev. A, Phys. Rev. A 65, 043805 (2002).

C Number of books or chapters submitted, but not yet published: 1

[1] R.Kolesov and O. Kocharovskaya, Mossbauer Spectroscopy, invited paper in UNESCO Enciclopedia, 2001.

D Number of books or chapters published (for each, provide a complete citation): 1

[1] A.B. Matsko, O. Kocharovskaya, Y. Rostovtsev, G.R. Welch, A.S. Zibrov, M.O. Scully, Slow, ultra-slow, stored and frozen light, The advances in Atomic, Molecular, and Optical Physics 46, 191 (2001), edited by B. Bederson and H. Walther.

E Number of printed technical reports/non-refereed papers (for each, provide a complete citation): 0

F Number of patents filed: 2

- Infrared generation in semiconductor lasers. Scully M.O., Belyanin A.A., Kocharovsky V.V. Patent Application No. TAMUS 1623 submitted to the U.S. Patent and Trademark Office on September 10, 2001, Serial No.: 09/950,458. Related International Patent Application: No. PCT/US01/28289.
- Detecting infrared radiation. Boyd R.W., Haden C.R., Scully M.O., Belyanin A.A., Kocharovsky V.V. Patent Application No. TAMUS 1750 submitted to the U.S. Patent and Trademark Office on October 31, 2001.

G Number of patents granted (for each, provide a complete citation): 0

H Number of invited presentations (for each, provide a complete citation): 25

- Workshop on Slow Light, Harvard, April 2000.

  1 Kocharovskaya O, Freezing light: Ultra-Slow Eit-Polariton with Vanishing or Negative Group Velocity

- Workshop on Quantum Nucleonics, Leuven (Belgium), May 2000.

  2 Kocharovskaya O., Laser control of Mossbauer transitions

- EOARD Workshop on Gamma-ray induced Transitions, London, May 2000.

  3 Kocharovskaya O, Problems and prospects for realization of gamma-ray laser

- International Conference "Mossbauer Effect: magnetism, modern materials, gamma optics", Kazan, July 2000:

  4 Kocharovskaya O, Laser manipulations by the Mossbauer transitions

- Sixteenth International Conference on Applications of accelerators in research and industry, Denton,Texas,November 2000,

5 O.Kocharovskaya, Laser manipulation by nuclear transitions

- 31st Winter Colloquium "Physics of Quantum Electronics", Snowbird, Utah, January 2001

6 Kocharovskaya O, Quantum coherence effects: solids vs gases

7 Belyanin A, Resonant parametric generation of infrared radiation on intersubband transitions in low-dimensional semiconductor heterostructures

8 Rostovtsev Y., Kocharovskaya O., Scully M.O., Freezing light via atomic motion

- All-Union Workshop "Nanophotonics 2001", Nizhny Novgorod, March 26-29 2001.

9 Belyanin A., Resonant parametric generation of infrared radiation on intersubband transitions in low-dimensional semiconductor heterostructures

- Symposium on coherent sources: semiconductor lasers and gamma-rays, April 4-6, 2001, Brussels, Belgium.

10 O. Kocharovskaya, Laser control of nuclear transitions as a way to laser-Mossbauer spectroscopy and gamma-ray laser

11 Y. Rostovtsev, Electromagnetically Induced Transparency for gamma-ray transition

- 17-th International Conference on Coherent and Nonlinear Optics ICONO, Belarus, Minsk (June, 2001),

12 Kocharovskaya O. and Kolesov R., Atomic interference phenomena: from optics to gamma-rays

- International Conference on Progress in Nonlinear Science, dedicated to the 100th Anniversary of A.A. Andronov, Nizhny Novgorod, Russia, July 2-6, 2001.

13 Kocharovskaya O, Nonlinear optics with coherently prepared medium

14 Belyanin A., Resonant parametric generation of infrared radiation on intersubband transitions in low-dimensional semiconductor heterostructures

- International Workshop on Laser Physics and Quantum Optics, Jackson Hole, Wyoming, July 29-August 4, 2001.

15 Belyanin A., Coherent radiation from neutral molecules moving above the grating

16 Rostovtsev Y., EIT for nuclear transition via level crossing

- XXII Solvay Conference in Physics, November 2001, Delphi, Greece. Invited talk.

17 Kocharovskaya O., Nuclear Interference Effects for Quantum Information Processing,

- International Conference on Lasers 2001, Tucson, Arizona (2001),

18 Kocharovskaya O., Rostovtsev Y., Kolesov R., Mossbauer Gamma-Ray Laser with Optical Driving

19 Rostovtsev Y., Kolesov R.. Kocharovskaya O., Laser-Mossbauer Spectroscopy as a New Tool for Nuclear Transitions

20 Coussement R, EIT for gamma-radiation

- 32nd Winter Colloquium "Physics of Quantum Electronics", Snowbird. Utah, January 2002.

21 Kocharovskaya O., Electromagnetically induced transparency in gamma-rays

22 Belyanin A., Transistor laser for wave mixing

23 Rostovtsev Y., Kocharovskaya O., Scully, Stop and go control of light Pulse via quantum coherence

24 Kolesov R., Optical continua generation in a coherently prepared Raman medium, oral

25 E. Kuznetsova, O. Kocharovskaya, P. Hemmer, and M. O. Scully. Atomic interference phenomena in solids with a long-lived spin coherence

I Number of submitted presentations (for each, provide a complete citation): 17

- International Conference "Mossbauer Effect: magnetism, modern materials, gamma optics", Kazan, July 2000:

  1 Kolesov R., Internal electronic conversion due to neighboring atoms, oral

- American Physical Society Texas Section Meeting, April 2000, College station, Texas, USA.

  2 Kocharovskaya O, Quantum interference effects: gases vs solids

  3 Kolesov R., The resolution of the gamma-ray laser dilemma

  4 Rostovtsev Y., Kocharovskaya O., Scully M.O., Stopping light in hot gases

- CLEO/Europe-IQEC, International Quantum Electronics Conference IQEC'00, Nice, France, September 10-15, 2000.

  5 Y.V. Radeonychev and Olga Kocharovskaya, Laser control of decay rates in three-level atoms

  6 O. Kocharovskaya, R. Kolesov, and Y. Rostovtsev, Resolution of gamma-ray laser dilemma

  7 O. Kocharovskaya, A. Matsko, Y. Rostovtsev, and M. O. Scully, Lasing without inversion via decay induced coherence

  8 O. Kocharovskaya, Y. Rostovtsev, and M. O. Scully, Freezing light: ultraslow EIT polariton in a three-level mediu

  9 O. Kocharovskaya, R. Kolesov, Y. Rostovtsev, and A. Belyanin, Laser manipulation of Mossbauer transitions as a novel method of gamma-ray spectroscopy

- 9th International Symposium "Nanostructures: Physics and Technology", S.-Petersburg, Russia, June 18-22, 2001 (2 papers).

  10 Belyanin A., Infrared generation in low-dimensional semiconductor heterostructures via quantum coherence

- 7th International Symposium on the Photon Echo and Coherent Spectroscopy, Novgorod, Russia, June 20-24, 2001.

  11 Belyanin A., Infrared generation in low-dimensional semiconductor heterostructures via quantum coherence

- International Workshop "Mid-Infrared Coherent Sources", S.-Petersburg, Russia, June 25-29, 2001 (1 paper).

  12 Belyanin A., Inversionless lasing with self-generated driving field

- 5th All-Union Conference on the Physics of Semiconductors. Moscow, September 10-14, 2001.

  13 Belyanin A., Coherent radiation from neutral molecules moving above the grating

- International Conference on Progress in Nonlinear Science, dedicated to the 100th Anniversary of A.A. Andronov, Nizhny Novgorod, Russia, July 2-6, 2001.

  14 E. Kuznetsova, R. Kolesov, and O. Kocharovskaya, Atomic interference phenomena in solids with a long-lived spin coherence,

- 17-th International Conference on Coherent and Nonlinear Optics ICONO, Belarus, Minsk (2001).

  15 Kolesov R. and Kocharovskaya O., Short pulse generation due to coherent population trapping

16 E. Kuznetsova, O. Kocharovskaya, and M. O. Scully, Electromagnetically induced transparency in solids with long-lived spin coherence,

17 Kolesov R., Kuznetsova E., Modification of Mossbauer spectra by means of polarization-selective optical pumping

J Honors/Awards/Prizes for contract/grant employees (list attached): 5

1. Olga Kocharovskaya, the PI of the contract, has given a tenure and has been promoted to Full Professor of Physics at Texas A&M University.
2. Zameer Hasan, the Co-PI has been promoted to the rank of Full Professor
3. Jeff Carroll, the Co-PI has been promoted to the rank of Full Professor
4. Roman Kolesov, PhD student, who is involved in this project, received an award of the International Conference on Mossbauer Spectroscopy for the best student's oral paper, and the TAMU Physics Graduate Student's Award for Excellence in Research.
5. The results obtained in [O.Kocharovskaya, Y.Rostovtsev, M.O.Scully, Freezing light via atomic coherence, Phys.Rev.Lett., 86, 628 (2001)] produced a great impact, and some articles discussing these results appeared in Physics Today [March 2001], Science News 155 (March 27), and Science Daily Magazine [http://www.sciencedaily.com/releases/2001/02/010201072301.html].

K Total number of full-time equivalent graduate students and postdoctoral associates supported during this period, under this PR number: 2

Graduate Students: 2
Postdoctoral Associates: 0
including the number of
Female Graduate Students: 1
Female Postdoctoral Associates: 0
the number of
Minority* Graduate Students: 0
Minority* Postdoctoral Associates: 0
including the number of
Asian Graduate Students: 0
Asian Postdoctoral Associates: 0

L Other funding (list agency, grant title, amount received this year, total amount, period of performance and a brief statement regarding the relationship of that research to your ONR grant)

- Texas Advanced Technology and Research Programs, Eletcromagnetically induced transparency and lasing without inversion in rare-earth ions doped compounds, $184,950 (2000-2001).
- AFOSR, Laser Modification of Mossbauer Spectra in Eu2+:CaS, $30,000 (April-October 2001).
- DARPA, Laser Gamma-ray laser with coherent optical driving, $282,576 (2001-2002).
- Texas Advanced Technology and Research Programs, Multiple Raman Scattering in solids for the new coherent sources of ultrashort pulses, $150,000 (2002-2003).

Part II

(a1) Principal Investigator:
Olga Kocharovskaya
Department of Physics, Texas A&M University; College Station, TX 77843-4242

(a2) Co-Investigator::
Zameer Hasan Department of Physics, Temple University, Philadelphia, PA 19122

(a3) Co-Investigator::
James Carroll

Department of Physics and Astronomy, Center for Photon-Induced Processes X-Ray Effects, Youngstown State University, One university Plaza, Youngstown, Ohio 44555-3616

(b) Current Telephone number:
(979)845-2012

(c) Cognizant ONR Program Officer:
Frank Narducci

(d) Program Objective:
This project is directed to the development of the rigorous theory of the coherent laser control of gamma-ray nuclear transitions and making a proof-of-principle experiment demonstrating (i) the strong influence of laser radiation on gamma-ray nuclear transitions, (ii) the coherent effects in the gamma-ray range, (iii) the efficient laser-assisted Mossbauer spectroscopy of nuclei.

(e) Significant Results:

Modifications of Mossbauer spectra

i) **EIT for gamma-rays** In the first time electromagnetically-induced transparency (EIT) for gamma-rays at the nuclear transitions has been demonstrated by studying Mössbauer spectra of ^{57}Fe in a $FeCO_3$ crystal in the presence of a magnetic field. The experimental results have been explained in terms of quantum interference involving nuclear level-crossing.

ii) **Mossbauer spectra of 0.01% Eu^{2+}, Eu^{3+} in MgS** Eu^{2+} and Eu^{3+} Mössbauer spectra in MgS powder with the record small density of Eu ions of 0.01 atomic % These spectra provide a unique information about the ratio of Eu^{2+}/Eu^{3+} in MgS which is an important characteristics of this permanent hole-burning material. The density was chosen to be the same as in in Eu:MgS films which are produced at Temple University. So this experiment presents an important step forward a realization of laser control of nuclear transition because it shows that recording of Mossbauer spectra with such low density (which presently available in films) is possible.

iii) **Nuclear interference effects for quantum information processing**

Collective excitations of nuclear spins in solids manipulated by coherent optical pulses are proposed as a system of choice for implementation of quantum information processing. It is shown that a high speed of the optical excitation of nuclear polarization (of the order of submicrosecond) can be nicely combined with a long storage time (of the order of second) and with a nearly 100% efficiency.

iv) **Laser-Mossbauer Spectroscopy**

A new type of spectroscopic technique, Laser-Mossbauer spectroscopy, which is based on comparison of the original Mossbauer spectra with those modified under the action of laser radiation was suggested. This technique, providing a unique information about nuclear structure and crystal environments which is unavailable by any other methods, is illustrated by calculation of possible laser modifications of Mossbauer spectra for a number of different nucleads.

v) **Mossbauer Gamma-ray laser with coherent optical drive**

The novel concept of Mossbauer gamma-ray laser with coherent optical drive is suggested. The essence of this concept is in usage of the resonant optical driving of bounded electrons in atoms in order to provide a suppression of resonant absorption by 3-5 orders of magnitude reducing it up to the level of the off-resonant losses in the host matrix. It leads to two profound consequences: (i) release in requirement for incoherent pump threshold and accordingly in the heating of the active sample, providing the possibility for pumping of active nuclei in the host lattice without destroying the conditions of both Mossbauer and Borrmann effects (ii) possibility to pump directly at the operating nuclear transition, increasing additionally the efficiency of the incoherent pump by 1-2 orders of magnitude.

Electromagnetically induced transparency(EIT) and its applications

i) **Atomic interference phenomena in solids with a long-lived spin coherence** We generalize a theory of EIT and slow group velocity for the case of inhomogeneouse line broadening on both one and two-photon- transitions which unavoidably takes place in solid materials with a long-lived spin coherence. We identify the new regimes of EIT where EIT linewidth is proportional to the amplitude of the driving field and where it can be essentially reduced due to inhomogeneous broadening of an optical transition. We suggest also a new class of solid materials, namely, rare-earth ions doped semi-conductors or dielectrics with electro-dipole allowed transitions, which is very promising for realization and applications of EIT.

ii) **Ultra-short pulse generation in solid medium with long-lived spin coherence** We show that utmost possible Raman coherence between equally populated levels can be excited by two-photon resonant bichromatic field far from one-photon resonance with an optical line via relaxation process. On this basis we suggest a novel mechanism for generation of extremely short pulses with high pulse duty factor and large energy in the pulse. It can be realized in solid materials, such as rare-earth doped dielectrics with long-lived spin coherence or NV-centers in diamond and provide a simple , reliable and convenient technique for many practical applications

iii) **Mixed electromagnetically and self-induced transparency**

We show that application of self-induced transparency (SIT) solitons as a driving field in V-type electromagnetically induced transparency (EIT) leads to "mixed induced transparency" (MIT) that nicely combines the best features of both SIT and EIT.

Ultraslow and stopped light and its applications

i) **Stopping light via hot atoms**

We prove that it is possible to freeze a light pulse (i.e., to bring it to a full stop) or even to make its group velocity negative in a coherently driven Doppler broadened atomic medium via electromagnetically induced transparency (EIT). This remarkable phenomenon of the ultraslow EIT polariton is based on the spatial dispersion of the refraction index n(w,k), i.e., its wave number dependence, which is due to atomic motion and provides a negative contribution to the group velocity. This is related to, but qualitatively different from, the recently observed light slowing caused by large temporal (frequency) dispersion.

ii) **Nonadiabatic approach to quantum optical information storage** We show that there is no need for adiabatic passage in the storage and retrieval of information in the optically thick vapor of lambda-atoms. We show that even in the case of instantaneous switching of the writing and reading fields compared with adiabatic switching, an almost perfect information storage is possible if the group velocity of the signal pulse is much less than the speed of light in the vacuum and the bandwidth of the signal pulse is much less then the width of the two photon resonance.

iii) **Multiplexing, time-reversing and transporting of the light via atomic coherence** We show that manipulating with the characteristics of the control pulses allows to transfer the information about the quantum state of the signal pulse (stored in the medium) to the light pulse with different frequency,

the state of polarization, or the direction of propagation than original signal pulse. It can be called multiplexing stored light. Even the time-reversing of the pulse becomes possible: so that its tail can precede to the head and vice versa. Moreover, the coherently driven atoms moving in the gas can transport the quantum information from one spatial point to another (transporting stored light). The experiments in support of our theoretical consideration have been performed in Rb vapor.

iv) **Using slow light to enhance acousto-optical effects: Application to squeezed light and anomalous stimulated Brillouin scattering**

We propose a technique for achieving phase matching in Brillouin scattering in a dielectric fiber doped by three-level Lambda-type ions. This can lead to a dramatic increase of efficiency of ponderomotive nonlinear interaction between the electromagnetic waves and holds promise for applications in quantum optics such as squeezing and quantum nondemolition measurements.

We study stimulated Brillouin scattering (SBS) in an ultradispersive coherent medium, and show that the properties of SBS change drastically when the group velocity of light in the material approaches or becomes less than the speed of sound. In particular, forward SBS not allowed in a dispersionless bulk medium takes place in the coherent medium.

New types of lasers without inversion

i) **Infrared generation in low-dimensional semiconductor heterostructures via quantum coherence**

A scheme for infrared generation without population inversion between subbands in quantum-well and quantum-dot lasers is presented. The scheme is based on the resonant nonlinear mixing of the optical laser fields on the two interband transitions that are generated in the same active region and that serve as the coherent drive for the infrared field. This mechanism for frequency down-conversion does not rely upon any ad hoc assumptions of long-lived coherences in the semiconductor active medium, and it should work efficiently at room temperature with injection current pumping. For optimized waveguide and cavity parameters, the intrinsic efficiency of the down-conversion process can reach the limiting quantum value corresponding to one infrared photon per one optical photon. Due to the parametric nature of infrared generation, the proposed inversionless scheme is especially promising for long-wavelength (far-infrared) operation.

ii) **Lasing without inversion via decay-induced coherence**

Continuous wave lasing without population inversion based on interference of the spontaneous coupled with field reservoir, in the absence of any coherent driving fields. Practical implementation of the scheme based on a pseudophotonic band-gap structure doped by three-level ions or semiconductor quantum dots is suggesed.

iii) **Free-Electron Lasers without inversion: Gain optimization and implementation scheme**

We consider a scheme of two noncollinear wigglers with an intermediate magnetic drift region, constituting a free-electron laser without inversion (FELWI). Two mechanisms of phase shifts in the drift region between the wigglers owing to a series of magnetic lenses can give rise to FELWI: velocity- and angle-dependent shifts. An appropriate combination of these shifts is shown to provide the conditions for amplification without inversion.

We propose also experimentally simplified schemes of an optically dispersive interface region between two coupled free electron lasers (FELs), aimed at achieving a much broader gain bandwidth than in a conventional FEL or a conventional optical klystron composed of two separated FELs. The proposed schemes can enhance the gain of FELs, regardless of their design when operated in the short pulsed regime.

Impact:

i) First experimental demonstration of EIT in gamma-rays at the nuclear transitions

ii) New methods for generation of gamma-ray and infrared radiation

iii) A novel configuration of free-electron laser without inversion

iv) New type of spectroscopy, Laser-Mossbauer spectroscopy.

v) New regimes of EIT (EIT line-narrowing, mixed EIT-SIT) and new EIT media (E1 electronic and nuclear transitions in rare-earth doped crystals)

vi) New regimes for quantum information storage (nonadiabatic storage, multipleaxing, time-reversing and transporting stored light)

vii) A dramatic enhancement of nonlinear magneto-optic effects via atomic coherence

viii) New method of ultra-short pulse generation in a solid medium with a long-lived coherence

ix) Proposal for full stopping the light

x) Proposal for usage of nuclear spins in rare-earth doped solids as a system of choice for implementation of quantum information processing

Summary of Plans

Further theoretical investigation and experimental demonstration of

i) Nuclear spectra manipulations by means of laser radiation: observation of HF structure from the optically excited electronic transitions and suppression of the resonant gamma-ray absorption

ii) EIT and ultra-slow group velocity for gamma-rays at the nuclear transitions

iii) Ultra-short pulse generation in solids with a long-lived spin coherence

iv) Infrared generation via quantum coherence

Detailed description and status of the experimental research on "Modification of Mössbauer Spectra by Laser Radiation"

Summary

This part of report describes the experimental research effort in the past twelve months to optimize the performance parameters of rare earth doped solid state materials, particularly in regard to atomically engineering large size samples that can be used to strongly couple the radiation field of a laser to the electronic states of a Mössbauer active rare earth ion. Such materials are potential candidates for coherently controlling the nuclear states of a Mössbauer active ion utilizing the hyperfine coupling. These research efforts were motivated by the past successes of atomic engineering of rare earth doped alkaline earth sulfides for ultra high–density and fast spectral storage where a similar atomic engineering of materials was required.

In this report results are presented for three different stages of experimental efforts for developing and testing new and improved materials for Mössbauer –Optical Double Resonance: 1. Thin films of Europium doped alkaline earth sulfides have been fabricated using Laser Pulsed Vapor Deposition Technique. Atomic parameters for Eu, have been optimized for a strong electron–radiation coupling. This is because Eu^{151} is also Mössbauer active and well suited for Mössbauer –Optical Double Resonance 2. Optical studies have confirmed that, in principle, the strongest possible coupling of the radiation field, of any solid–state system, can be achieved for the Eu electronic transitions in alkaline earth sulfides. 3. Mössbauer studies have been performed on these atomically engineered systems. These studies confirm that Eu–doped sulfides are ideally suited for observing the effects of coherent coupling of the laser radiation with the nuclear states of Eu using Mössbauer effect.

Plans are also presented for the next phases in experimental and theoretical research so as to ultimately demonstrate the coherent control of nuclear states.

I. Introduction

In the past year, we have worked on a program to develop and study materials where the nuclear states of an optically active ion can be coherently controlled using an intense optical field. A laser provides the optical field. It coherently controls the electronic transitions that in turn control the nuclear states by hyperfine coupling between the electronic and nuclear states. The manifestation of such a coherent control of nuclear states would be in the form of new lines or modified line shapes of the hyperfine structure observed in the Mössbauer spectrum[1].

Coherent control of nuclear states has many possible exciting applications in defense, new technology, basic science, medicine, and biology:

- Lasing without inversion in the γ–ray range, i.e., the realization of a γ–ray laser.
- Nuclear polarization by optical pumping
- Electromagnetically induced transparency in the γ–ray range.
- Direct measurement of nuclear radii
- Precise measurement of isomer shifts.
- An opportunity to study the hyperfine contribution of a particular electron from the localized outer shell

Naturally, the focus of our investigation is on solid–state materials embedded with optically active ions that exhibit strongly allowed electronic transitions so as to efficiently couple them to the radiation field. At the same time, the nuclei of these ions should exhibit a strong Mössbauer effect.

Tailoring of such materials is not an easy task. It requires a delicate balance of the macroscopic properties of the host medium and the microscopic or atomic properties of the optically active ion[2].

The experimental effort of the project was divided into four stages:

1. Atomic tailoring of suitable materials that have potential to exhibit strong electron–radiation coupling in Mössbauer active atoms or ions.

2. Optical studies of the above atomically tailored materials to confirm their strong electron–radiation coupling.
3. Mössbauer studies of the above materials to confirm their suitability for the double resonance experiments.
4. Mössbauer –Optical Double Resonance (MODOR) studies by combining the Mössbauer and optical experiments.

In the first year of the program we engineered the atomic scale properties by doping Mössbauer active impurities in II–VI wide bandgap materials. Testing their optical characteristics demonstrated that europium doped alkaline earth sulfides in general, and magnesium and calcium sulfides in particular, have very desirable parameters for the proposed experiments. The tailoring of strongly allowed electronic transitions in the red region of the spectrum, the lack of host nuclear spins which prolongs coherence, and the possibility of designing these materials in the form of thin films, are collectively responsible for such extraordinary preference for these materials.

In the Eu–doped II–VI materials the oscillator strengths of optical transitions can be maximized to a value approaching that of the strongest allowed atomic transitions in a solid. The naturally occurring isotope of Europium, Eu^{151} (natural abundance ~50%) is Mössbauer active. Therefore, optical manipulation of the Eu nuclear states can be investigated by Mössbauer –optical double resonance. In this proposed technique, a high power laser excites Eu ions to their excited electronic state while changes in the Mössbauer spectrum are monitored as a result of this excitation.

The main limitation of these materials is that they are very difficult to grow in the form of single crystals. Only one example of a single crystal growth (of poor optical quality) exists in literature[3]. Alkaline earth sulfides are usually grown in the form of polycrystalline powder that is unsuitable for any application where large size samples with high optical quality are needed.

In the first part of the project, the main emphasis of the program was to develop the materials in two– or three–dimensional form suitable for the proposed investigations. Two–dimensional thin films of these materials were prepared by Laser Pulsed Vapor Deposition (LPVD). A controlled fabrication of these films yielded samples that exhibited strongly absorbing Zero–Phonon Lines of Eu^{2+}.

Almost in parallel to our efforts of material preparation, the program of developing experimental facilities for Mössbauer studies on Eu was initiated. The set–up for the Mössbauer studies has been successfully used for observing Eu spectra. It will be described in detail later in section IV.

As can be seen, in the first year of the research activity the first two stages of the project were carried out in parallel with the third stage.

II. Material Requirements

Material requirements for samples suitable for MODOR can be divided into two parts:

A. Requirements for Electronic Energy Levels

1. High oscillator strength for the optical transition for a strong coupling of the radiation field (for Eu 4f–5d.
2. 2. Small inhomogeneous linewidth of the transition so as to be pumped efficiently by a laser with narrow line–width._
3. A weak Electron– Phonon coupling. It broadens the electronic absorption lines. Therefore, only low temperature experiments are possible. A small Debye–Waller factor is also desirable for the observation of strong Mössbauer effect.
4. A weak ion–ion interaction. It gives many undesirable effects. Low concentration of optically active impurity can suppress such effects. However, it also results in a weak Mössbauer signal
5. No Host Nuclear Spins. Otherwise spin–spin interactions may cause undesirable decoherence effects.
6. Transition wavelength or driving optical frequency should lie in the range where commercial tunable lasers operate, $\lambda \sim 550–700$ nm.

4f–5d transitions of Eu^{2+} satisfy most of the above requirements in A. To tailor the energies of this transition in the right wavelength range ($\lambda \sim 550–700$ nm) Eu doped in II–VI sulfides form the ideal systems. As discussed below Eu^{151} is also suitable as a Mössbauer active isotope.

B. Requirements for Observing Strong Mössbauer Effect

1. High Probability of Mössbauer effect. As a Mössbauer active isotope Eu^{151}, is the third best after Fe^{57} and Sn^{119}. Unlike Eu^{2+} the last two do not have a strong and sharp electronic transition in a solid host in the visible range
2. High abundance of Mössbauer isotope. Mössbauer active isotope Eu151 is about 50% abundant
3. High $T_{1/2}$ (half life) of Mössbauer nuclide after Fe, Eu has the longest, ~10 ns lifetime.
4. Available commercial radioactive source with strong Mössbauer effect at room temperature.

Combining the requirements in A and B seriously limits the choices of solid host–ion combination for MODOR studies.

III. Objectives

Faced with the task of designing and fabricating the materials as well as optimizing their optical and Mössbauer properties, the following were the objectives of our research activity in the past year:

1. Identification of Solid State Materials for γ– ray–optical double resonance.
2. Fabrication of the materials identified, in particular, thin films of Eu–doped II–VI sulfides.

3. To improve upon the optical properties of the laser pulsed vapor deposited thin films of CaS:Eu and MgS:Eu for high optical quality and small inhomogeneous broadenings. This was achieved by controlling the growth conditions for the films.
4. Characterization of mechanical, optical and atomic properties of CaS:Eu and MgS:Eu films.
5. Designing and testing of experiments for Mössbauer studies of CaS: Eu and MgS:Eu.
6. Mössbauer studies of Eu in MgS and CaS hosts.

IV. Accomplishments

Our research accomplishments in the past year are summarized below. A more detailed account of the projects follows this summary.

A. Atomic Engineering of Solids

We have identified and engineered materials, Eu doped II–VI sulfides particularly MgS:Eu and CaS:Eu, that have the following attractive features simultaneously:

- Electronic transitions with the highest oscillator strength in a solid. 4f–5d transition of Eu^{2+} is electric dipole allowed with $f \sim 0.01$ and therefore is ideal for strong radiation–electron coupling.
- Right frequency range for optical transition to be manipulated by a laser (λ = 625nm and 575nm for CaS:Eu and MgS:Eu respectively).
- Eu^{151} is one of the best possible Mössbauer active nucleus.
- In most systems there are no host nuclear spins to adversely affect the coherence. In CaS natural isotopes of both Ca and S have zero nuclear spin. Nuclear spins are zero for a great fraction of naturally abundant isotopes of Mg (90%), Sr (93%) and Ba (83%) as well. Therefore if need be. their, isotopically pure samples are relatively easy to obtain.
- Strong Hyperfine Interaction in the ground and excited states to satisfy selection rules for coherent excitation.

B. Fabrication and Preparation of Samples

As we prepare all of our samples and the starting material in house the accomplishment have been in the following areas:

- Preparation of powder samples of CaS:Eu and MgS:Eu using high temperature chemistry and thermal diffusion of Eu.
- Thin films of CaS and MgS by pulsed laser deposition.
- Atomic tailoring of films to optimize Eu electronic absorption and other parameters.

Achievements for making the ultimate samples in the form of thin films are as follows:

1. Thin films of CaS and MgS doped with Eu have been fabricated. The ratio of Eu^{2+} to Eu^{3+} has been optimized to increase the concentration of Eu^{2+}–optical centers. The inhomogeneous width of the 4f–5d transition has been reduced to enhance coupling with the laser.
2. Optical investigations have confirmed that for the Laser Pulse Vapor Deposited films exhibit high oscillator strength of the Eu^{2+} transition and mostly a single Eu–center is present in the samples.
3. These films are typically a few microns thick. Fabrication of thick samples is not possible using this technique. However this is not a limitation. Several layers of high quality thin films over each other can be used to form a thick sample. Therefore, a few microns thick multi–layer stack could provide adequate concentration for significant absorption of gamma rays in Mössbauer experiments.

C. Mössbauer Investigations

In parallel with fabrication and optical studies of materials, in the past year we have also started Mössbauer investigation. Following are the accomplishments for Mössbauer studies:

1. Designing and setting up a Mössbauer laboratory with a Co^{57} source for testing purposes and a Sm^{151} source for Mössbauer investigations of Eu^{151}.
2. Recording of MgS:Eu Mössbauer spectrum using the facilities developed. Such studies have also helped in identifying the concentration ratio of Eu^{2+} / Eu^{3+}. A feedback of this information to the materials fabrication is highly valuable in order to maximize Eu^{2+} concentration in the samples.
3. A very significant achievement has been in reducing the time of accumulation of Mössbauer data. For dilute samples, Eu ~ 0.01 molar concentration, it was estimated to be about two months. It has been demonstrated that by improving the detection techniques good Mössbauer spectra can be accumulated in a week to ten days time for such dilute samples.

In the following we give more details of the experiments.

i). Materials Processing and Preparation (M.F. Aly and Z. Hasan)

As we have mentioned above, all materials that we study are prepared in our laboratories. Facilities used for the present investigations included micro-particle preparation lab and the thin film fabrication laboratory.

Micro-particles of MgS and CaS with Eu were prepared by reducing the respective sulfates in the presence of a reducing atmosphere at high temperature, close to 1000 ^{0}C. A desired concentrations of Eu_2O_3 in the starting mixture controlled the Eu concentration. Under high temperature Eu diffuses in the lattice to replace Mg or Ca to form the Eu^{2+} and Eu^{3+} centers. Details of the preparation of these samples are described elsewhere[i].

ii). Laser Pulse Vapor Deposition (LPVD) of Thin Films of MgS:Eu and CaS:Eu

(M.F. Aly, F. Vagizov and Z. Hasan)

Thin films in large sizes with right atomic properties are very attractive candidates for the proposed experiments of MODOR. The fabrication of thin films of MgS and CaS doped with Eu was started last year. These films were prepared using our home built LPVD system shown in Figure 1. It is based on a XeF excimer laser operating at 350 nm. An ultrahigh vacuum chamber equipped with multiple-target and substrate holders is the core of the setup. In this chamber, the chemical environment during the growth is adjustable by introducing the reactive, oxidizing or reducing, gases. This control allowed for the enhancement of the concentration of Eu^{2+} at the cost of Eu^{3+} in the films.

Targets of MgS and CaS doped with the desired concentration of Eu were prepared by high pressure compressing of micro-particles followed by a high temperature annealing cycle. These targets were also prepared in our laboratories.

Although the sulfide lattice prefers Eu^{2+} state a high fraction of Eu can be in the form of Eu^{3+}, which is also very stable in the lattice. By optimizing the growth conditions, such as the temperature of growth, the pressure of the reactive gases in the chamber, laser pulse frequency, and laser power we have greatly enhanced the Eu^{2+} concentration in our samples and hence the performance of thin MgS:Eu and CaS:Eu films for MODOR experiments[4,5], (Figure 2).

Thin films formed in our experiments are highly glassy in nature and therefore the ZPLs are generally broad. The inhomogeneous broadening of the purely electronic transition, known as Zero Phonon Line (ZPL) can also be controlled by the temperature of deposition. At high temperatures of growth the optical quality of the sample is good at the same time the inhomogeneous widths are greatly reduced (Figure 3). All optical measurements were performed in Dr. Hasan's High Resolution Laser Laboratory at Temple University. With reduced linewidths of ZPL a strong coupling of the laser with the electronic states of Eu is possible.

iii). Mössbauer studies of Eu in MgS and CaS. (Temple University: F. Vagizov, A. Konjhodzic, and Z. Hasan, Youngstown State University: Jeff Carroll)

During the last year a new laboratory was established for Mössbauer studies pertaining to these investigations at Temple University. As no equipment purchase was possible in fiscal planning, this lab was built with internal resources and some starting equipment available from one of the investigator's (Jeff Carroll) laboratory.

The experimental set up developed in our laboratory is shown in Figure 4. Mössbauer spectra of MgS:Eu were recorded using this setup. Several modifications such as the use of scintillation counter rather than a proportional counter, stabilization of room temperature to within 1^0C, the use of highly stabilized signal amplifiers, etc., were needed to record the spectrum in Figure 5. It clearly shows Mössbauer signals associated with Eu^{2+} and Eu^{3+} in a sample of MgS with 0.01 molar percentage of Eu. This was a powder sample and the data was accumulated over a period of five days. This is a very significant result for the MODOR experiment that has to be performed on thin films of high optical quality so as to be able to optically excite the Eu ions.

Prior to the experiment shown in Figure 5 it was calculated that a relatively high concentration of centers (~10^{19}per cubic centimeter) was needed for the detection of a good Mössbauer spectrum. Increasing the concentration has serious implications for the electronic states. At high concentrations many different Eu centers can be formed due to ion–ion interactions. Therefore, a low concentration of Eu in the sample is a necessary requirement. For such dilute samples the estimated data accumulation time was two months. Data in Figure 5 confirms that no more than a week of data accumulation is needed to record a Mössbauer spectrum with good signal to noise ratio[6].

Mössbauer studies of similarly dilute CaS:Eu samples have not been encouraging up to this stage.

IV. Status of Effort

The current status of the research for the coherent control of nuclear states can be summarized as follows:

1. Thin films of CaS and MgS doped with Eu have been grown by laser pulsed vapor deposition. These films are suitable for the MODOR experiments. Optical quality of the film and relative concentration of Eu^{2+}/ Eu^{3+} have been improved by controlling growth conditions such as the temperature, atmosphere, and the rate of growth. Both MgS and CaS films have some attractive features. CaS films are generally of higher optical quality their ZPL's are narrow but they have a weaker oscillator strength, *f*, for the 4f–5d transition of Eu^{2+}. MgS:Eu has strong oscillator strength for the transition. Its ZPL is broad in comparison to CaS :Eu, and therefore it would be less efficiently coupled to the laser field.
2. Mössbauer signal is easily detectable from dilute MgS:Eu samples in the form of micro–particles. In comparison CaS:Eu does not show a significant signal even for a five fold increase in the data accumulation times. The only advantage of CaS over MgS is the absence of host spins. However, if

MgS:Eu samples are prepared with isotopes of Mg for which (I=0), this problem can be solved. Almost 90% of naturally occurring Mg has zero nuclear spin. Such isotopically pure samples would be ideally suited for MODOR experiments.

3. Although theoretical estimates show that with low concentration of Eu, 1mm thick samples are needed for a decent signal to noise ratio in the Mössbauer spectrum. Such spectrum will need the same, one–week accumulation time. However, this thickness requirement can be greatly reduced in the following manner:

    - Using only Eu^{151} in MODOR samples that is almost 51% abundant in the current samples. Therefore, for isotopically pure samples the thickness requirements are reduced from 1mm to 500 μ.
    - Observing Mössbauer effect in constant velocity mode will reduce the number of velocity channels to be monitored by a factor of 10 from the present case where the spectrum is monitored in constant acceleration mode. This will reduce the thickness to 50 μ.
    - A 10 μ thick stack of thin films will require only 5–10 times greater accumulation time. This falls within the reasonable limit of 5–10 weeks for MODOR experiment.

In conclusion, in the past year we have removed a large number of barriers toward the realization of a solid–state material that can be considered suitable for coherent control of nuclear states using a laser. In light of this progress, MODOR experiments on Eu–doped II–VI materials look very promising and should be continued.

IV. Future Directions

Future research efforts would focus on the following three areas in a timely manner:

i) Materials preparation and optical characterization

- Preparation and fabrication of powder and thin films with single isotope Eu^{151} samples.
- To Suppress Eu^{3+} and maximize the Eu^{2+} concentration in MgS and CaS samples
- Optical characterization of powder and thin films of MgS:Eu, and CaS:Eu

ii) Mössbauer studies

- Optimization of Sm^{151} Mössbauer spectra of Eu in MgS and CaS films.

iii) Mössbauer –Optical Double Resonance studies

- Setting up the double resonance experiment with Sm^{151} source for Mössbauer of $MgS:Eu^{151}$ and $CaS:Eu^{151}$. The experimental set up for MODOR is the same as in Figure 4 with the exception that the optical excitation of the sample is done by an Ar–ion pumped dye laser and changes in the Mössbauer spectrum are monitored upon optical excitation.

- The development of the software for computer control of MODOR experiment. This is particularly needed as the anticipated experiments require long times for the data acquisition extending to several weeks.

V. Personnel Supported

(a). Temple University

Faculty

1. Dr. Zameer Hasan, Co–Principal Investigator, no financial support for the academic year or summer salaries.

Post–Doctoral Fellows

1. Dr. Farit Vagizov, part time
2. Dr. Moniruzzaman, part time

Graduate students

1. Mr. Aras Konjdodzic, part time
2. Mr. Sameh Dardona, part time

References:

1. Olga Kocharovskaya, Phys.Rev.Letters 82, 3593 (1999)
2. Z. Hasan, L. Biyikli, and P. I. Macfarlane, "Power–Gated Spectral Hole–burning in an Unstable System ($MgS:Eu^{2+},Eu^{3+}$): A Case for High Density Gated Hole–burning". App. Phys. Letters, 72, 3399–3401 (1998).
3. Y. Kaneko and T. Koda "New Developments in IIa –VIb (Alkaline–Earth Chalcogenide) Binary Semiconductors", Journal of Crystal Growth 86,72–78 (1988)
4. Z.Hasan, International Conference on Physics of Quantum Electronics, Snowbird, UT. Jan 2001 Title: Thin Films of CaS:Eu and MgS:Eu for Electronic and Nuclear Coherences.

5. Aras Konjhodzic, F. Vagizov and Z. Hasan, International Conference on Physics of Quantum Electronics, Snow Bird, UT, Jan. 2002 Title: Coherent Control of Nuclear States: Mössbauer Studies of Eu^{2+}/Eu^{3+} in MgS
6. F. Vagizov, Aras Konjhodzic and Z. Hasan, Mössbauer Studies of Eu^{2+}/Eu^{3+} in MgS:Eu thin films deposited by Laser Pulsed Vapor Deposition, (to appear in Journal of Modern Optics)

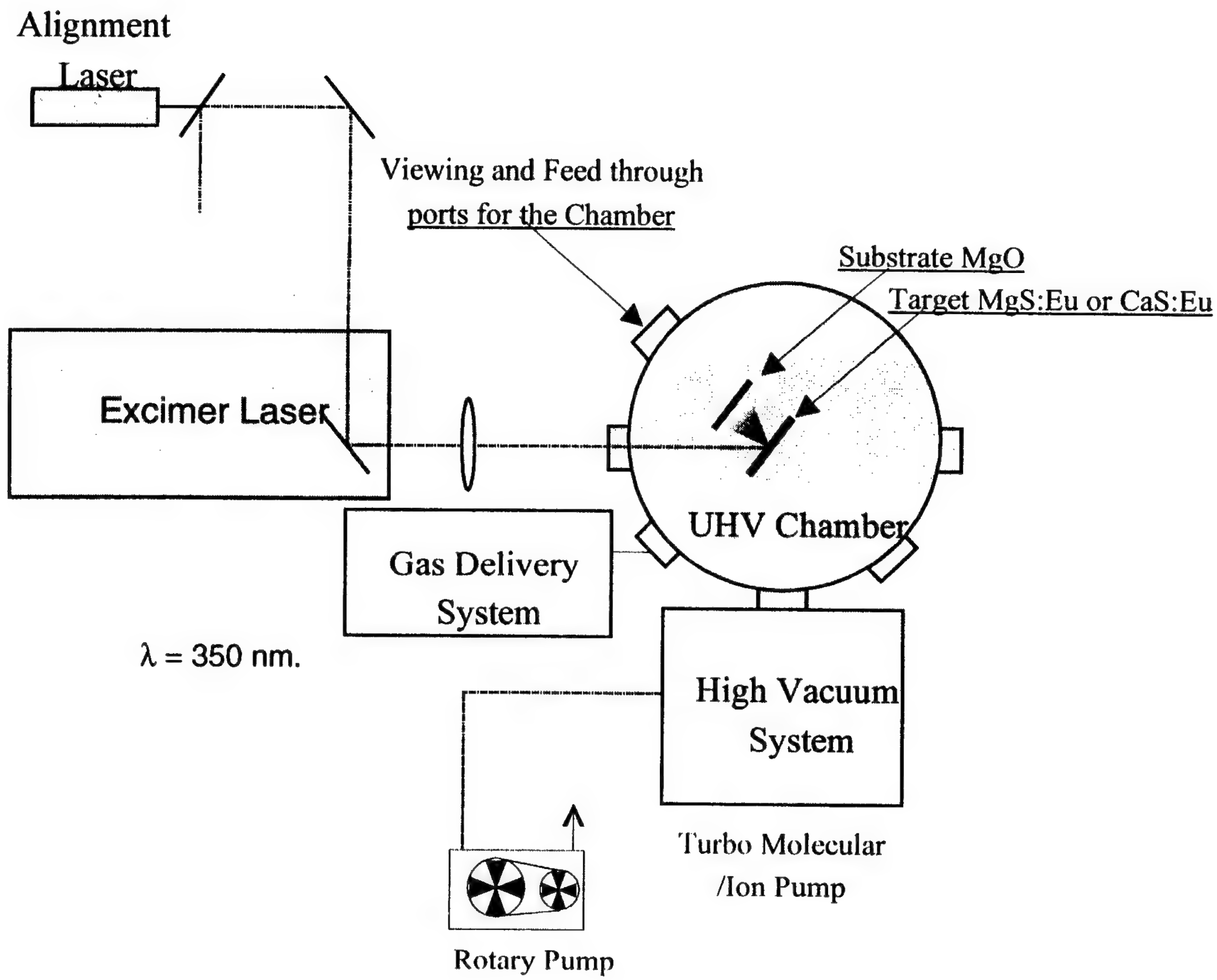

Figure 1. LPVD Setup for thin film deposition

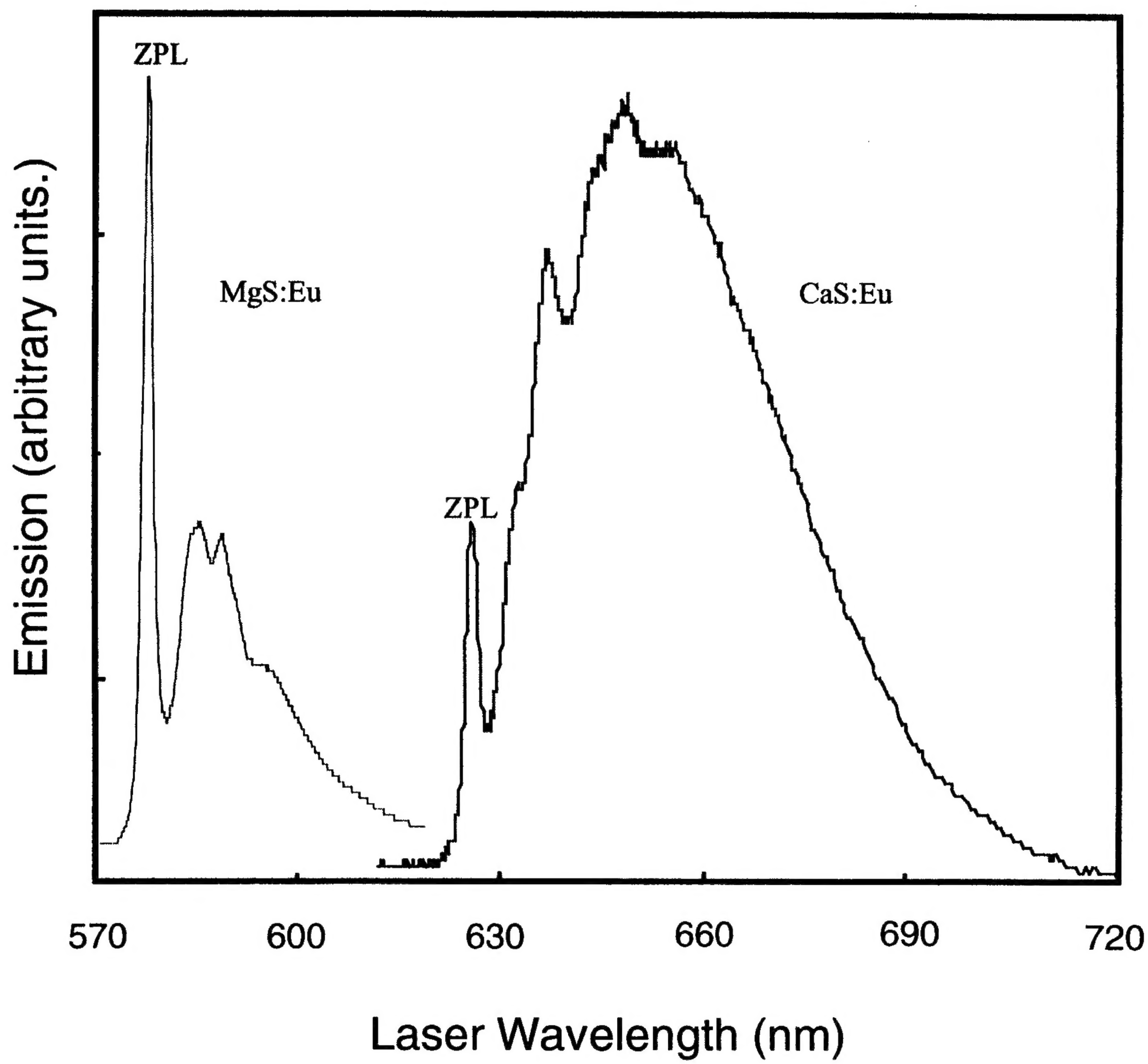

Figure 2. Emission spectra of CaS:Eu and MgS:Eu thin films.

Sharp electronic transitionsare indicated by ZPLs.

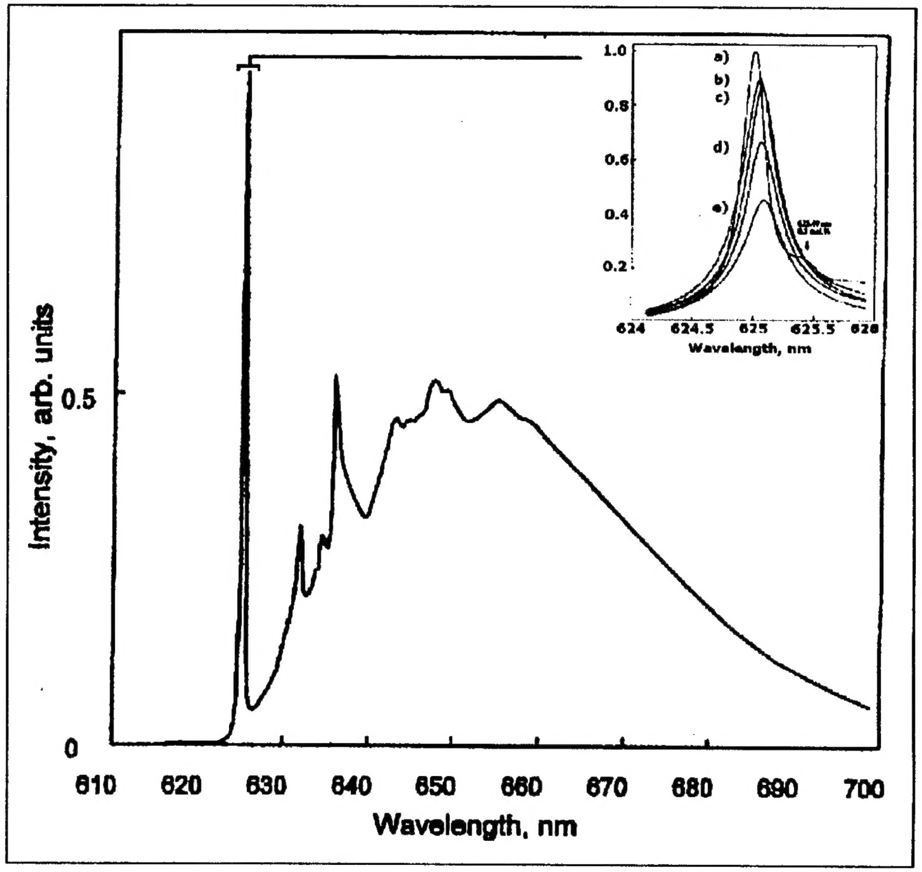

Fig. 3. The emission ZPL in CaS:Eu as a function of the concentration of Eu. ***a)*** **0.2,** ***b)*** **0.05,** ***c)*** **0.01,** ***d)*** **0.08,** ***e)*** **0.004 molar %.**

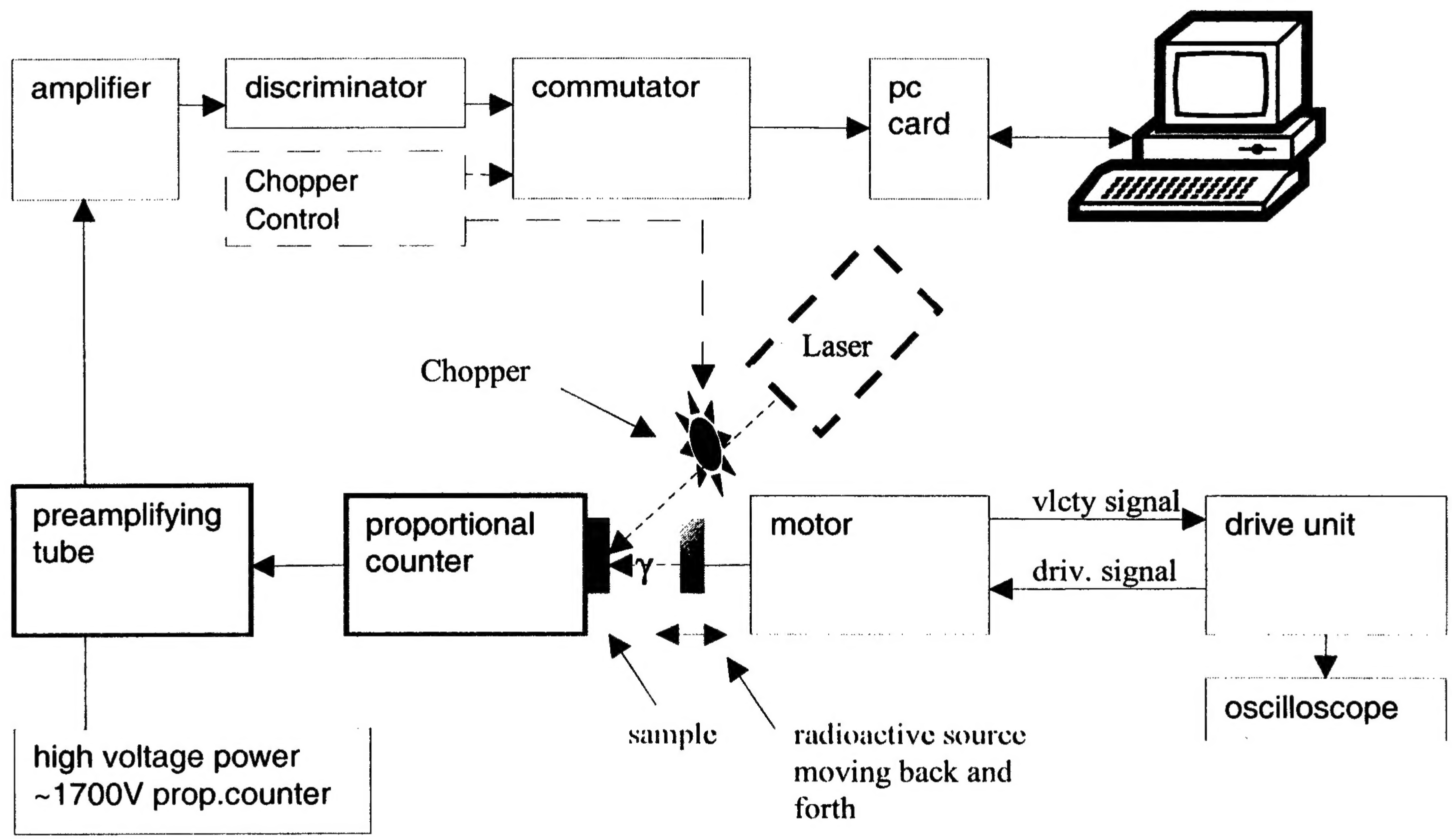

Figure 4. Mossbauer Apparatus in Constant Velocity Mode. MODOR set up requires additional components indicated by dotted lines

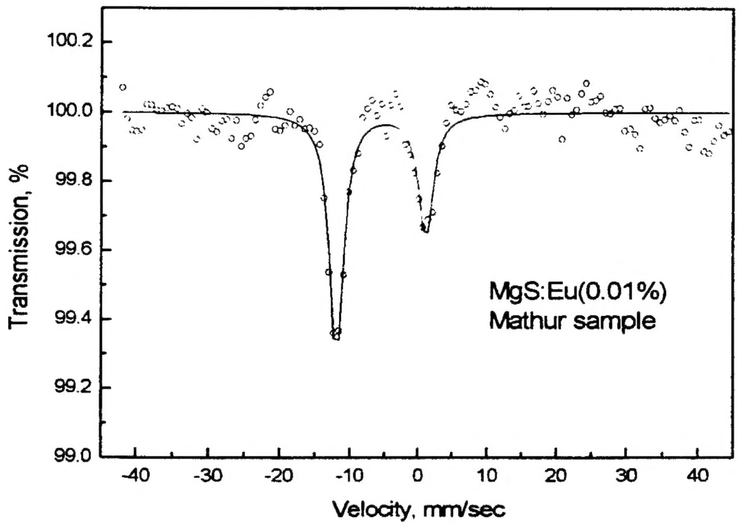

Figure 5. MØssbauer Spectrum of MgS:Eu indicating the ratio, Eu^{3+}/Eu^{2+}.